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CHAPTER II

Preliminary Classification:
Proposed Class:
Subclass:

TRANSMITTAL LETTER TO THE UNITED STATES ELECTED OFFICE (EO/US)

(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)

EP00/07937	15 August 2000 (15.08.00)	17 September 1999 (17.09.99)
International Application No.	**International Filing Date**	**Priority Date Claimed**

TITLE OF INVENTION: OPTICAL METHOD FOR CHARACTERIZATION OF PARTICULATE SYSTEMS AND DEVICE FOR CARRYING OUT SAID METHOD

APPLICANT(S): APSYS ADVANCED PARTICLE SYSTEMS GmbH; VALET, Oliver K.; GUENTHER, Peter A.

Box PCT
Assistant Commissioner for Patents
Washington D.C. 20231
ATTENTION: EO/US

CERTIFICATION UNDER 37 C.F.R. ' ' 1.8(a) and 1.10*
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TRANSMISSION

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** Only the date of filing (' 1.6) will be the date used in a patent term adjustment calculation, although the date on any certificate of mailing or transmission under ' 1.8 continues to be taken into account in determining timeliness See ' 1 703(f) Consider "Express Mail Post Office to Addressee" (' 1 10) or facsimile transmission (' 1 6(d)) for the reply to be accorded the earliest possible filing date for patent term adjustment calculations*

1. Applicant herewith submits to the United States Elected Office (EO/US) the following items under 35 U.S.C. § 371:

   a. This express request to immediately begin national examination procedures (35 U.S.C. § 371(f)).

   b. The U.S. National Fee (35 U.S.C. § 371(c)(1)) and other fees (37 C.F.R. § 1.492) as indicated below:

2. Fees

CLAIMS FEE*	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
	TOTAL CLAIMS	14 -20 =	0	x $18.00 =	$0.00
	INDEPENDENT CLAIMS	2 - 3 =	0	x $84.00 =	$0.00
	MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ $280.00	$0.00
BASIC FEE	U.S. PTO WAS NOT INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where no international preliminary examination fee as set forth in § 1.482 has been paid to the U.S. PTO, and payment of an international search fee as set forth in Section 1.445(a)(2) to the U.S. PTO: where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 C.F.R. § 1.492(a)(5)) $890.00				$890.00
	Total of above Calculations				= $890.00
SMALL ENTITY	Reduction by 1/2 for filing by small entity, if applicable. Assertion must be made. (note 37 C.F.R. § 1.27)				- $0.00
	Subtotal				$890.00
	Total National Fee				$890.00
	Fee for recording the enclosed assignment document $40.00 (37 C.F.R. § 1.21(h)). See Item 13 below). See attached "ASSIGNMENT COVER SHEET".				$0.00
TOTAL	Total Fees enclosed				$890.00

THE FEE FOR EXCESS CLAIMS, IF ANY, WILL BE PAID UPON THE FILING OF THE TRANSLATION OF THE SPECIFICATION.

Attached is a check in the amount of $890.00.

A duplicate copy of this sheet is enclosed.

3. A copy of the International application as filed (35 U.S.C. § 371(c)(2)) has been transmitted by the International Bureau. An additional copy is filed herewith.

Date of mailing of the application (from form PCT/IB/308): not known.

4. A translation of the International application into the English language (35 U.S.C. § 371(c)(2)) will follow.

5. Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. § 371(c)(3)) have not been transmitted. Applicant chose not to make amendments under PCT Article 19.

Date of mailing of Search Report (from form PCT/ISA/210): 9 January 2001.

6. A translation of the amendments to the claims under PCT Article 19 (38 U.S.C. § 371(c)(3)) has not been transmitted for reasons indicated at point 5(c) above.

7. A copy of the international examination report (PCT/IPEA/409) Transmitted herewith

8. Annex(es) to the international preliminary examination report is/are not transmitted herewith , as there are none to transmit.

9. A translation of the annexes to the international preliminary examination report is not transmitted herewith, as there is no annex to translate.

10. An unexecuted oath or declaration of the inventor (35 U.S.C. § 371(c)(4)) complying with 35 U.S.C. § 115 is submitted herewith, and such oath or declaration identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or 3(c) and 5(b); and states that they were reviewed by the inventor as required by 37 C.F.R. § 1.70.

II. Other document(s) or information included:

11. An International Search Report (PCT/ISA/210) or Declaration under PCT Article 17(2)(a) has been transmitted by the International Bureau.

Date of mailing (from form PCT/IB/308): 9 January 2001.

12. An Information Disclosure Statement under 37 C.F.R. §§ 1.97 and 1.98 will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. § 371(c).

13. Additional documents:

a. Copy of request (PCT/RO/101)

b. International Publication No. 01/22061
   i. Front page only

14. The above items are being transmitted before 30 months from any claimed priority date.

Date: 13 MARCH 2002

Signature of Practitioner

Reg. No.: 33390
Tel. No.: 330-864-5550
Customer No.: 021324

Stephen L. Grant
Hahn Loeser & Parks LLP
1225 W. Market St.
Akron, OH 44313
USA

Attorney's Docket 117040-57

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Valet, et al.	Examiner:	Unknown
Ser. No.:	10/088,008	Art Group:	Unknown
Title:	OPTICAL METHOD FOR CHARACTERIZATION OF PARTICULATE SYSTEMS AND DEVICE FOR CARRYING OUT THE METHOD		
Filed:	13 March 2002	Date:	18 June 2002

PRELIMINARY AMENDMENT

This Preliminary Amendment is filed with the filing of the missing parts in the above case, which claims priority from an application filed in Germany as 101 10 358.1 on 27 February 2001. The fees for the claims should be calculated based on the claims remaining after the entry of this Preliminary Amendment, which results in 26 total and 3 independent claims. Because the initial specification filed was in German and a verified translation is provided herewith, a substitute specification under 37 CFR §1.125 is provided instead of a clean copy of the amendments made below.

Amendments to the Disclosure

The specification as filed has been altered from the literal translation document received to delete information above the title, to insert headings according to US practice, and to insert paragraph numbering in lieu of line numbering. No new matter has been added.

Amendments to the Abstract

Please amend the Abstract as follows:

An [The invention concerns an] optical method and an associated device is provided for characterization of particulate systems, with which the particles present in a particulate system, for example a clean room, can be detected in respect of quantity and size and at the same time information about the identity of the particles can be provided. An [In accordance with the invention an] air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is

then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light.
The [In accordance with the invention] identification of the particles is effected by combined laser-Raman spectroscopy which [, with a short exposure time, by virtue of the use of powerful light sources, strong-light optics and in particular by the renunciation of high levels of optical resolution,] affords results which can be used for automated evaluation. The low level of spectral resolution is used to advantage.

Amendments to the Drawings

A substitute Figure 2 is presented to translate German legends in to English, as supported by the verified translation.

Amendments to the Claims

After the heading "CLAIMS" and before the beginning of the claims, please insert the words: -- What is claimed is: --

Please amend the claims as follows:

1. (amended) An optical method for characterization of particulate systems, wherein [characterized in that] an air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light.

2. (amended) The [A] method of claim 1, wherein [as set forth in claim 1 characterized in that] the scattered light measuring unit triggers the optical system of the identification unit by means of a control.

3. (amended) The [A] method of claim 2, wherein [as set forth in claim 2 characterized in that] particles with preselected properties are investigated by means of logical linking of the scattered light measuring unit and the optical system of the identification unit.

4. (amended) The [A] method of claim 3, wherein [as set forth in one of claims 1 through 3 characterized in that] identification of the particles is effected by means of combined laser-Raman spectroscopy.

5. (amended) The [A] method of claim 4, wherein [as set forth in one of the preceding claims characterized in that] the particle is reduced to such a speed that a measuring time of between approximately 1 ms and 1 s is available for the particle.

6. (amended) The [A] method of claim 5, wherein [as set forth in one of the preceding claims characterized in that] the Raman spectra obtained are compared after chemometric analysis with a database and associated therewith.

7. (amended) A device for carrying out a method of optically characterizing particulate systems in a flow of an ambient air, [the method as set forth in one of claims 1 through 6] comprising:

a particle feeder and

an electronic evaluation unit for characterizing particulate systems, comprising at least the following modules [characterized in that the unit for characterization of particulate systems comprises module units which include at least]:

[-] an optical unit for determining the size and number of particles in the [an] air flow [from the ambient air],

[-] a particle brake,

[-] an optical identification unit for the moved particles contained in the air flow, comprising corona discharge, excitation laser and spectrometer units [unit], and

[-] an electronic control.

8. (amended) The [A] device of claim 7, wherein [as set forth in claim 7 characterized in that] the particle brake is an electromagnetic brake.

9. (amended) The [A] device of claim 8, wherein [as set forth in claim 7 or claim 8 characterized in that] the optical identification unit comprises [includes] a narrow-band light source and an NIR multichannel spectrometer.

10. (amended) The [A] device of claim 9, wherein [as set forth in claim 9 characterized in] the light source is a monochromatic light source.

11. (amended) The [A] device of claim 10, wherein [as set forth in one of claims 7 through 10 characterized in that] the spectrometer unit comprises at least one microspectrometer.

12. (amended) The [A] device of claim 11, wherein said at least one [as set forth in claim 11 characterized in that] the microspectrometer is arranged in such a way to achieve [that] spectral resolution of at least fifteen wave numbers [is achieved].

13. (amended) The [A] device of claim 11, wherein [as set forth in one of claims 7 through 12 characterized in that] the microspectrometer of the optical identification unit is replaced by other suitable spectroscopic devices in dependence on the particles to be analyzed.

14. (amended) The [A] device of claim 12, wherein [as set forth in one of claims 7 through 13 characterized in that] the electronic control comprises [is an electronic control which includes] a programmable AD-converter card with integrated processor and an integrated control program.

Please add the following new claims:

15. (new) The device of claim 7, wherein the optical identification unit comprises a narrow-band light source and an NIR multichannel spectrometer.

16. (new) The device of claim 15, wherein the light source is a monochromatic light source.

17. (new) The device of claim 7, wherein the spectrometer unit comprises at least one microspectrometer.

18. (new) The device of claim 7, wherein the electronic control comprises a programmable AD-converter card with integrated processor and an integrated control program.

19. (new) An optical method for the characterizing a particulate system in a flow of an ambient air, the method comprising the steps of:

guiding at a defined speed the air flow containing particles from the ambient air by a particle feeder past a first scattered light measuring unit;

detecting light scattered by the particles in the first measuring unit:

reducing the speed of the particles; and

identifying the reduced speed particles in the air flow in an identification unit by means of interaction with monochromatic light.

20. (new) The method of claim 19, wherein the step of detecting scattered light in the scattered light measuring unit triggers a control in the identification unit.

21. (new) The method of claim 20, wherein the particles having preselected properties are investigated by means of the combination of the scattered light measuring unit and the optical system of the identification unit.

22. (new) The method as of claim 21, wherein the identifying step is effected by a combined laser-Raman spectroscopy.

23. (new) The method of claim 22, wherein the speed reducing step reduces the particle to such a speed to provide a measuring time of between 1 millisecond and 1 second.

24. (new) The method of claim 22, wherein the identifying step further comprises comparing and associating the Raman spectra obtained after chemometric analysis with a database.

25. (new) The method as of claim 19, wherein the identifying step is effected by a combined laser-Raman spectroscopy.

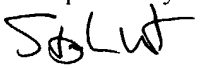
26. (new) The method of claim 19, wherein the speed reducing step reduces the particle to such a speed to provide a measuring time of between 1 millisecond and 1 second.

REMARKS

The above claims have been amended to more closely correspond them to United States claiming practice, namely, by removing multiple dependencies, especially improper multiple dependencies, by removing reference numerals, and by clarifying antecedent basis issues. These amendments to the claims are fully supported by the literal translation into English of the specification as filed in Germany, and they do not introduce new subject matter.

None of the above claims are narrower than the multiple dependent claims from which they are drawn and represent only content already claimed in the specification as originally filed.

Respectfully submitted,



Stephen L. Grant
Reg. No. 33,390
Hahn Loeser & Parks LLP
1225 W. Market St.
Akron, OH 44313
330-864-5550
Fax 330-864-7986
Email: Grant@oldhamlaw.com
Customer No. 021324

OPTICAL METHOD FOR CHARACTERIZATION OF PARTICULATE SYSTEMS AND DEVICE FOR CARRYING OUT THE METHOD

[0001] The invention concerns an optical method for characterization of particulate systems, in particular for clean room monitoring, with which the particles present in a particulate system, for example a clean room, can be detected in respect of quantity and size and at the same time information about the identity of the particles can be provided. The invention also concerns a device for carrying out the method. The method and the associated device make it possible for example to implement preventative quality assurance in clean rooms.

Background of the Art

[0002] Because of the increasing miniaturization of products, the electronics industry involves very high demands in terms of the purity and cleanliness of the gases involved in the production process. If the product structures are of the same order of magnitude as the diameters of the particles to be encountered in the gases, they act as "killer particles" in the production process. The cleanliness demands in terms of the room air in the production processes in the electronics industry are therefore becoming more and more strict and require knowledge about the origin, movement and substance composition of the particles. At the present time in principle two different methods are used for particle measurement and particle analysis separately from each other.

[0003] So-called particle counters are used for determining the level of particle concentration in the clean room air. These involve measuring devices which are capable of continuously measuring an air sample from the clean room. These can be measuring devices which by way of an optical method can detect particles to a size of 0.1 μm and can attribute them to given size categories. Special particle counters, so-called condensation nucleus counters, permit the measurement of particles to a size of 0.05 μm. That is made possible by virtue of the fact that particles are increased in size due to condensation of a liquid and are measured thereafter.

[0004] The particle counters serve exclusively for counting the particles, and analysis of the material composition of the particles is not possible. The measured

particles are also no longer available for later analysis with other measurement systems as the sample volume is abandoned after flowing through the measuring device. It is therefore necessary to take another sample again, for further analysis procedures.

[0005] There are numerous measuring devices for particle analysis, which make it possible to establish the material composition of particles. Those measuring devices operate inter alia on the basis of the principle of electron-laser spectroscopy. The measurement systems are generally disposed in separate laboratories because generally they are not suitable for a clean room situation and they require very difficult sample preparation. Direct analysis of the particles in the clean room air is not possible with those devices.

[0006] At the present time there is a method in the course of development, which can analyze both the number and size of particles and also the particle composition. That method is based on the mass-spectroscopic analysis of particles which were ionized by means of UV-lasers. That technology however is not suitable for a clean room situation by virtue of the oil pumps used. In addition the size of the measuring unit does not permit mobile use thereof and it will probably be very expensive.

[0007] The object of the present invention is to develop a method and an associated device for detecting in respect of quantity and size and simultaneously determining the identity of the particles present in a particulate system, in particular in a clean room, which permits automatic online operation, which is suitable for a clean room situation, which is inexpensive and industry-standardized and which can be put to mobile use.

Summary of the Invention

[0008] In accordance with the invention that object is attained by an optical method for characterization of particulate systems as set forth in claim 1 and a device for carrying out the method as set forth in claim 7.

[0009] In accordance with the invention an air flow comprising the ambient air is guided at a defined speed through a particle feeder past a first scattered light

measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed is identified in the air flow in an identification unit by means of interaction with monochromatic light.

[0010] It is possible for the first time with that method to quantitatively detect the particles present in a clean room and at the same time to provide information about the identity of the particles. This therefore provides the clean room operators with a tool which for the first time makes it possible to implement preventative quality assurance and thus to very substantially satisfy the rising demands in terms of cleanliness of the room air used in the electronic production process.

[0011] In a preferred configuration of the invention the optical system of the identification unit, the spectrometer unit, is triggered by way of an electronic control by the scattered light measuring unit. By virtue thereof, it is possible in the analysis step following the particle size determining operation, if required, to identify only particles in a preselected range, that is to say for example only particles of a given diameter or in a given diameter range.

[0012] The selection criteria can be determined and selected under software control by means of the electronic control. Such a selection option is particularly advantageous in terms of using the identification unit in particle-rich environments.

[0013] In accordance with the invention, identification of the particles is effected by combined laser-Raman spectroscopy which, with a short exposure time, by virtue of the use of high-power light sources, strong-light optics and in particular by virtue of foregoing high levels of optical resolution (normally 4 cm^{-1}, here 12-24 cm^{-1}), affords results which are usable for automated evaluation. The low level of spectral resolution is used to advantage.

[0014] The reduction of the speed of the particles to a residence time of about 1 ms to about 1 s in the second laser beam serves to obtain vibration spectra which show all spectral features and which are suitable for automated evaluation. Signals which are obtained without the speed reduction of the particle are not sufficient for identification in the predominant number of cases as the noise increases greatly and therefore automated evaluation of the spectra becomes impossible.

[0015] The Raman spectra obtained are electronically filtered and investigated for spectral features (peaks) and the peak table obtained is finally compared to a database which contains corresponding reference tables and the substance identified.

[0016] The device according to the invention comprises module units which include at least the following elements:

- an optical unit for determining the size and number of particles in an air flow from the ambient air,
- a particle brake,
- an optical identification unit for the moved particles contained in the air flow, comprising corona discharge, excitation laser and spectrometer unit, and
- an electronic control.

[0017] The modular structure of the system is an essential point of view as on the one hand it permits further development and application in wider areas of use while on the other hand it permits the replacement of individual modules by other suitable ones according to the properties of the particles to be identified. Thus for example different demands can be made on the spectrometer unit, depending on whether organic impurities or biotic particles have to be identified. For example, a resonance Raman module would be used for the identification of biotic particles, either jointly with the Raman module or instead of the Raman module.

[0018] In addition the system is preferably in the form of a mobile unit, involving dimensions of a maximum of about 1 x 2 x 1 m and of a weight of about 40 kg so that use can take place directly at the location to be sampled and the samples do not have to be sent to analysis laboratories. That for example makes it possible to implement preventative clean room monitoring.

[0019] The light source of the identification unit is preferably a narrow-band light source, preferably a monochromatic light source. The spectrometer unit of the identification unit is preferably formed from an NIR multichannel spectrometer. The multichannel spectrometer preferably has about 255 detectors and preferably has a measuring range of approximately 900 – 1,900 nm. That technology is inexpensive and permits the desired small dimensions for the entire measuring device.

[0020] By virtue of the only short available measuring times of between 1 ms and 1 s, particular demands are to be made on the light source. A particularly suitable light source has been found to be a narrow-band light source, preferably a monochromatic light source with a high power output. It is however also possible to use other suitable laser light sources, for example multimode laser diodes, wide-band laser diodes and pulsed laser light sources.

[0021] By virtue of the renunciation of resolution and the use of simple components, this new technology combines laser spectroscopy with the simplicity and convenience of other optical methods, for example NIR spectroscopy. In contrast to FT spectroscopy that permits the very short measurement times specified.

[0022] The electronic control, on the basis of predetermined parameters such as for example the size of the particle, after interaction with the first scattered light measuring unit, decides whether the particle is or is not analyzed in the identification unit. For that purpose, reading-out is effected by way of a programmable AD-converter card with integrated processor, preferably an 80x86 processor, at a frequency of about 20 kHz, the size or the refractive index is ascertained by means of the integrated program and compared to the preset value. If the particle falls in the range which is of interest, a trigger signal is sent to the identification unit, whereupon the particle is characterized.

[0023] Use of the integrated AD-converter card gives a very high level of system safety and security. Use of the electronic circuit basically permits use of the described system in particle-rich environments in which the identification unit, without preselection, would be overloaded.

Brief Description of the Drawings

[0024] The invention will be described hereinafter by means of an embodiment. In the accompanying drawings:

[0025] Figure 1 is a diagrammatic view of the module units and the co-operation thereof, and

[0026] Figure 2 shows vibration spectra involving conventional Raman technology and NIR-Raman technology according to the invention.

[0027] The method and the device according to the invention which is used for carrying out the method are to be presented by way of example in terms of the identification of a polymer microparticle of a size of between 0.5 μm and 15 μm, which is typical for contamination in clean rooms.

[0028] Contamination in a clean room, for example of the 1,000 category, is sucked in by a pump and converted by means of a nozzle and a particle feeder 1 into an individual particle flow.

[0029] In this situation the particle 10 generally acquires a speed of approximately 10 mm/s. That particle 10 now passes the first laser beam L1 which is emitted for example by an HeNe-laser 2 with approximately 20 mW output power and focused at 50 μm. The scattered light is detected in dependence on angle and evaluated to determine the particle size in accordance with the known theory of elastic light scatter (Mie theory). If a selection of given particles is to be effected, for example in accordance with a given diameter, the laser 2, upon fulfillment of the selection properties by the detected particle, sends a trigger signal by way of a control 3 to the downstream-connected identification unit which comprises a corona discharge 4, an excitation laser 5 and a spectrometer unit 6. The selectable properties of the particles can be preselected by means of software at the electronic control 3. That selection technology is particularly advantageous in regard to use of the identification unit in particle-rich environments. If no selection is to be effected the trigger signal is sent at each detected particle 10.

[0030] After detection and determination of size by the first laser 2 the particle 10 passes the corona discharge 4 which is operated for example at 10,000 V. In this case the particle 10 is occupied with charge proportionally to the surface area. In a downstream-disposed electromagnetic field, a so-called electromagnetic brake 7, the particle 10 is decelerated to a speed of about 1 mm/s, thus affording a resonance time of the particle 10 in the second laser beam L2 of about 10 ms. The laser beam L2 is preferably produced by a semiconductor laser 5 with a wavelength of 780 nm and an output power of 300 mW and focused to 10 μm beam diameter.

[0031] The light which is inelastically scattered in that period is detected after suppression of the excitation wavelength by means of a holographic notch filter by

between one and three minispectrometers 6, the geometrical arrangement of which is such that spectra are obtained with a resolution of 12 cm^{-1} over a wavelength range of 200 – 4,000 cm^{-1} relative to the excitation wavelength. The vibration spectrum obtained in that way in the range of 200 - 4,000 cm^{-1} is electronically filtered and investigated for spectral features (peaks).

[0032] The peak table obtained is finally compared to a database 8 which contains the necessary reference tables and the substance of the particle 10 identified.

[0033] Figure 2 shows the spectrum of a palmitic acid particle which was used as a test system. It is of a diameter of about 4 μm and was recorded once with a residence time in the second laser beam L2 of 10 ms and once with a shorter residence time, that is to say without braking of the particle 10. The lower spectrum shows the recording with conventional Raman technology and the upper spectrum shows that with a short exposure time of 10 ms and a resolution of 12 cm^{-1} with the procedure according to the invention on an individual particle. With a short exposure time, all spectral features can be recognized, and the noise is markedly more pronounced.

[0034] In the predominant number of cases, the signal which is obtained without use of the electromagnetic brake, that is to say without a prolongation of the available measuring time, is not sufficient for identification as the noise increases greatly and therefore automated evaluation becomes impossible.

CLAIMS

What is claimed is:

1. An optical method for characterization of particulate systems, wherein an air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light.

2. The method of claim 1, wherein the scattered light measuring unit triggers the optical system of the identification unit by means of a control.

3. The method of claim 2, wherein particles with preselected properties are investigated by means of logical linking of the scattered light measuring unit and the optical system of the identification unit.

4. The method of claim 3, wherein identification of the particles is effected by means of combined laser-Raman spectroscopy.

5. The method of claim 4, wherein the particle is reduced to such a speed that a measuring time of between approximately 1 ms and 1 s is available for the particle.

6. The method of claim 5, wherein the Raman spectra obtained are compared after chemometric analysis with a database and associated therewith.

7. A device for carrying out a method of optically characterizing particulate systems in a flow of an ambient air, comprising:

a particle feeder and

an electronic evaluation unit for characterizing particulate systems, comprising at least the following modules:

an optical unit for determining the size and number of particles in the air flow,

a particle brake,

an optical identification unit for the moved particles contained in the air flow, comprising corona discharge, excitation laser and spectrometer units, and

an electronic control.

8. The device of claim 7, wherein the particle brake is an electromagnetic brake.

9. The device of claim 8, wherein the optical identification unit comprises a narrow-band light source and an NIR multichannel spectrometer.

10. The device of claim 9, wherein the light source is a monochromatic light source.

11. The device of claim 10, wherein the spectrometer unit comprises at least one microspectrometer.

12. The device of claim 11, wherein said at least one the microspectrometer is arranged in such a way to achieve spectral resolution of at least fifteen wave numbers.

13. The device of claim 11, wherein the microspectrometer of the optical identification unit is replaced by other suitable spectroscopic devices in dependence on the particles to be analyzed.

14. The device of claim 12, wherein the electronic control comprises a programmable AD-converter card with integrated processor and an integrated control program.

15. The device of claim 7, wherein the optical identification unit comprises a narrow-band light source and an NIR multichannel spectrometer.

16. The device of claim 15, wherein the light source is a monochromatic light source.

17. The device of claim 7, wherein the spectrometer unit comprises at least one microspectrometer.

18. The device of claim 7, wherein the electronic control comprises a programmable AD-converter card with integrated processor and an integrated control program.

19. An optical method for the characterizing a particulate system in a flow of an ambient air, the method comprising the steps of:

guiding at a defined speed the air flow containing particles from the ambient air by a particle feeder past a first scattered light measuring unit;

detecting light scattered by the particles in the first measuring unit:

reducing the speed of the particles; and

identifying the reduced speed particles in the air flow in an identification unit by means of interaction with monochromatic light.

20. The method of claim 19, wherein the step of detecting scattered light in the scattered light measuring unit triggers a control in the identification unit.

21. The method of claim 20, wherein the particles having preselected properties are investigated by means of the combination of the scattered light measuring unit and the optical system of the identification unit.

22. The method as of claim 21, wherein the identifying step is effected by a combined laser-Raman spectroscopy.

23. The method of claim 22, wherein the speed reducing step reduces the particle to such a speed to provide a measuring time of between 1 millisecond and 1 second.

24. The method of claim 22, wherein the identifying step further comprises comparing and associating the Raman spectra obtained after chemometric analysis with a database.

25. The method as of claim 19, wherein the identifying step is effected by a combined laser-Raman spectroscopy.

26. The method of claim 19, wherein the speed reducing step reduces the particle to such a speed to provide a measuring time of between 1 millisecond and 1 second.

Abstract

An optical method and an associated device is provided for characterization of particulate systems, with which the particles present in a particulate system, for example a clean room, can be detected in respect of quantity and size and at the same time information about the identity of the particles can be provided. An air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light. The identification of the particles is effected by combined laser-Raman spectroscopy which affords results which can be used for automated evaluation. The low level of spectral resolution is used to advantage.

IN THE UNITED STATES PATENT AND TRADE MARK OFFICE

VERIFICATION OF TRANSLATION

I, Michael Wallace Richard Turner, Bachelor of Arts, Chartered Patent Attorney, European Patent Attorney, of I Horsefair Mews, Romsey, Hampshire SO51 8JG, England, do hereby declare that I am conversant with the English and German languages and that I am a competent translator thereof;

I verify that the attached English translation is a true and correct translation made by me of the attached specification in the German language of International Application PCT/EP00/07937;

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: April 11, 2002

M W R Turner

Berlin	14th August 2000
Our ref:	AB1004 JKB/js
Applicants/proprietors:	APSsys Advanced Particle Systems GmbH
Office ref:	New application

APSys Advanced Particle Systems GmbH,
Ostendstrasse 25, D-12459 Berlin

Optical method for characterization of particulate systems and device for carrying out the method

The invention concerns an optical method for characterization of particulate systems, in particular for clean room monitoring, with which the particles present in a particulate system, for example a clean room, can be detected in respect of quantity and size and at the same time information about the identity of the particles can be provided. The invention also concerns a device for carrying out the method. The method and the associated device make it possible for example to implement preventative quality assurance in clean rooms.

Because of the increasing miniaturization of products, the electronics industry involves very high demands in terms of the purity and cleanliness of the gases involved in the production process. If the product structures are of the same order of magnitude as the diameters of the particles to be encountered in the gases, they act as "killer particles" in the production process. The cleanliness demands in terms of the room air in the production processes in the electronics industry are therefore becoming more and more strict and require knowledge about the origin,

movement and substance composition of the particles. At the present time in principle two different methods are used for particle measurement and particle analysis separately from each other.

So-called particle counters are used for determining the level of particle concentration in the clean room air. These involve measuring devices which are capable of continuously measuring an air sample from the clean room. These can be measuring devices which by way of an optical method can detect particles to a size of 0.1 μm and can attribute them to given size categories. Special particle counters, so-called condensation nucleus counters, permit the measurement of particles to a size of 0.05 μm. That is made possible by virtue of the fact that particles are increased in size due to condensation of a liquid and are measured thereafter.

The particle counters serve exclusively for counting the particles, and analysis of the material composition of the particles is not possible. The measured particles are also no longer available for later analysis with other measurement systems as the sample volume is abandoned after flowing through the measuring device. It is therefore necessary to take another sample again, for further analysis procedures.

There are numerous measuring devices for particle analysis, which make it possible to establish the material composition of particles. Those measuring devices operate inter alia on the basis of the principle of electron-laser spectroscopy. The measurement systems are generally disposed in separate laboratories because generally they are not suitable for a clean room situation and they require very difficult sample preparation. Direct analysis of the particles in the clean room air is not possible with those devices.

At the present time there is a method in the course of development, which can analyze both the number and size of particles and also the particle composition. That method is based on the mass-spectroscopic analysis of particles which were ionized by means of UV-lasers. That technology however is not suitable for a clean room situation by virtue of

the oil pumps used. In addition the size of the measuring unit does not permit mobile use thereof and it will probably be very expensive.

The object of the present invention is to develop a method and an associated device for detecting in respect of quantity and size and simultaneously determining the identity of the particles present in a particulate system, in particular in a clean room, which permits automatic online operation, which is suitable for a clean room situation, which is inexpensive and industry-standardized and which can be put to mobile use.

In accordance with the invention that object is attained by an optical method for characterization of particulate systems as set forth in claim 1 and a device for carrying out the method as set forth in claim 7.

In accordance with the invention an air flow comprising the ambient air is guided at a defined speed through a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed is identified in the air flow in an identification unit by means of interaction with monochromatic light.

It is possible for the first time with that method to quantitatively detect the particles present in a clean room and at the same time to provide information about the identity of the particles. This therefore provides the clean room operators with a tool which for the first time makes it possible to implement preventative quality assurance and thus to very substantially satisfy the rising demands in terms of cleanliness of the room air used in the electronic production process.

In a preferred configuration of the invention the optical system of the identification unit, the spectrometer unit, is triggered by way of an electronic control by the scattered light measuring unit. By virtue thereof, it is possible in the analysis step following the particle size determining operation, if required, to identify only particles in a preselected range, that is to say for example only particles of a given diameter or in a given diameter range.

The selection criteria can be determined and selected under software control by means of the electronic control. Such a selection option is particularly advantageous in terms of using the identification unit in particle-rich environments.

In accordance with the invention, identification of the particles is effected by combined laser-Raman spectroscopy which, with a short exposure time, by virtue of the use of high-power light sources, strong-light optics and in particular by virtue of foregoing high levels of optical resolution (normally 4 cm^{-1}, here 12-24 cm^{-1}), affords results which are usable for automated evaluation. The low level of spectral resolution is used to advantage.

The reduction of the speed of the particles to a residence time of about 1 ms to about 1 s in the second laser beam serves to obtain vibration spectra which show all spectral features and which are suitable for automated evaluation. Signals which are obtained without the speed reduction of the particle are not sufficient for identification in the predominant number of cases as the noise increases greatly and therefore automated evaluation of the spectra becomes impossible.

The Raman spectra obtained are electronically filtered and investigated for spectral features (peaks) and the peak table obtained is finally compared to a database which contains corresponding reference tables and the substance identified.

The device according to the invention comprises module units which include at least the following elements:

- an optical unit for determining the size and number of particles in an air flow from the ambient air,
- a particle brake,
- an optical identification unit for the moved particles contained in the air flow, comprising corona discharge, excitation laser and spectrometer unit, and
- an electronic control.

The modular structure of the system is an essential point of view as on the one hand it permits further development and application in wider areas of use while on the other hand it permits the replacement of individual modules by other suitable ones according to the properties of the particles to be identified. Thus for example different demands can be made on the spectrometer unit, depending on whether organic impurities or biotic particles have to be identified. For example, a resonance Raman module would be used for the identification of biotic particles, either jointly with the Raman module or instead of the Raman module.

In addition the system is preferably in the form of a mobile unit, involving dimensions of a maximum of about 1 x 2 x 1 m and of a weight of about 40 kg so that use can take place directly at the location to be sampled and the samples do not have to be sent to analysis laboratories. That for example makes it possible to implement preventative clean room monitoring.

The light source of the identification unit is preferably a narrow-band light source, preferably a monochromatic light source. The spectrometer unit of the identification unit is preferably formed from an NIR multichannel spectrometer. The multichannel spectrometer preferably has about 255 detectors and preferably has a measuring range of approximately 900 – 1,900 nm. That technology is inexpensive and permits the desired small dimensions for the entire measuring device.

By virtue of the only short available measuring times of between 1 ms and 1 s, particular demands are to be made on the light source. A particularly suitable light source has been found to be a narrow-band light source, preferably a monochromatic light source with a high power output. It is however also possible to use other suitable laser light sources, for example multimode laser diodes, wide-band laser diodes and pulsed laser light sources.

By virtue of the renunciation of resolution and the use of simple components, this new technology combines laser spectroscopy with the simplicity and convenience of other optical methods, for example NIR

spectroscopy. In contrast to FT spectroscopy that permits the very short measurement times specified.

The electronic control, on the basis of predetermined parameters such as for example the size of the particle, after interaction with the first scattered light measuring unit, decides whether the particle is or is not analyzed in the identification unit. For that purpose, reading-out is effected by way of a programmable AD-converter card with integrated processor, preferably an 80x86 processor, at a frequency of about 20 kHz, the size or the refractive index is ascertained by means of the integrated program and compared to the preset value. If the particle falls in the range which is of interest, a trigger signal is sent to the identification unit, whereupon the particle is characterized.

Use of the integrated AD-converter card gives a very high level of system safety and security. Use of the electronic circuit basically permits use of the described system in particle-rich environments in which the identification unit, without preselection, would be overloaded.

The invention will be described hereinafter by means of an embodiment. In the accompanying drawings:

Figure 1 is a diagrammatic view of the module units and the co-operation thereof, and

Figure 2 shows vibration spectra involving conventional Raman technology and NIR-Raman technology according to the invention.

The method and the device according to the invention which is used for carrying out the method are to be presented by way of example in terms of the identification of a polymer microparticle of a size of between 0.5 μm and 15 μm, which is typical for contamination in clean rooms.

Contamination in a clean room, for example of the 1,000 category, is sucked in by a pump and converted by means of a nozzle and a particle feeder 1 into an individual particle flow.

In this situation the particle 10 generally acquires a speed of approximately 10 mm/s. That particle 10 now passes the first laser beam L1 which is emitted for example by an HeNe-laser 2 with approximately

20 mW output power and focused at 50 μm. The scattered light is detected in dependence on angle and evaluated to determine the particle size in accordance with the known theory of elastic light scatter (Mie theory). If a selection of given particles is to be effected, for example in accordance with a given diameter, the laser 2, upon fulfillment of the selection properties by the detected particle, sends a trigger signal by way of a control 3 to the downstream-connected identification unit which comprises a corona discharge 4, an excitation laser 5 and a spectrometer unit 6. The selectable properties of the particles can be preselected by means of software at the electronic control 3. That selection technology is particularly advantageous in regard to use of the identification unit in particle-rich environments. If no selection is to be effected the trigger signal is sent at each detected particle 10.

After detection and determination of size by the first laser 2 the particle 10 passes the corona discharge 4 which is operated for example at 10,000 V. In this case the particle 10 is occupied with charge proportionally to the surface area. In a downstream-disposed electromagnetic field, a so-called electromagnetic brake 7, the particle 10 is decelerated to a speed of about 1 mm/s, thus affording a resonance time of the particle 10 in the second laser beam L2 of about 10 ms. The laser beam L2 is preferably produced by a semiconductor laser 5 with a wavelength of 780 nm and an output power of 300 mW and focused to 10 μm beam diameter.

The light which is inelastically scattered in that period is detected after suppression of the excitation wavelength by means of a holographic notch filter by between one and three minispectrometers 6, the geometrical arrangement of which is such that spectra are obtained with a resolution of 12 cm^{-1} over a wavelength range of 200 – 4,000 cm^{-1} relative to the excitation wavelength. The vibration spectrum obtained in that way in the range of 200 - 4,000 cm^{-1} is electronically filtered and investigated for spectral features (peaks).

The peak table obtained is finally compared to a database 8 which contains the necessary reference tables and the substance of the particle 10 identified.

Figure 2 shows the spectrum of a palmitic acid particle which was used as a test system. It is of a diameter of about 4 μm and was recorded once with a residence time in the second laser beam L2 of 10 ms and once with a shorter residence time, that is to say without braking of the particle 10. The lower spectrum shows the recording with conventional Raman technology and the upper spectrum shows that with a short exposure time of 10 ms and a resolution of 12 cm^{-1} with the procedure according to the invention on an individual particle. With a short exposure time, all spectral features can be recognized, and the noise is markedly more pronounced.

In the predominant number of cases, the signal which is obtained without use of the electromagnetic brake, that is to say without a prolongation of the available measuring time, is not sufficient for identification as the noise increases greatly and therefore automated evaluation becomes impossible.

CLAIMS

1. An optical method for characterization of particulate systems, characterized in that an air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light.

2. A method as set forth in claim 1 characterized in that the scattered light measuring unit triggers the optical system of the identification unit by means of a control.

3. A method as set forth in claim 2 characterized in that particles with preselected properties are investigated by means of logical linking of the scattered light measuring unit and the optical system of the identification unit.

4. A method as set forth in one of claims 1 through 3 characterized in that identification of the particles is effected by means of combined laser-Raman spectroscopy.

5. A method as set forth in one of the preceding claims characterized in that the particle is reduced to such a speed that a measuring time of between approximately 1 ms and 1 s is available for the particle.

6. A method as set forth in one of the preceding claims characterized in that the Raman spectra obtained are compared after chemometric analysis with a database and associated.

7. A device for carrying out the method as set forth in one of claims 1 through 6 comprising a particle feeder and an electronic evaluation unit, characterized in that the unit for characterization of particulate systems comprises module units which include at least:

- an optical unit for determining the size and number of particles in an air flow from the ambient air,
- a particle brake,
- an optical identification unit for the moved particles contained in the air flow, comprising corona discharge, excitation laser and spectrometer unit, and
- an electronic control.

8. A device as set forth in claim 7 characterized in that the particle brake is an electromagnetic brake.

9. A device as set forth in claim 7 or claim 8 characterized in that the identification unit includes a narrow-band light source and an NIR multichannel spectrometer.

10. A device as set forth in claim 9 characterized in the light source is a monochromatic light source.

11. A device as set forth in one of claims 7 through 10 characterized in that the spectrometer unit comprises at least one microspectrometer.

12. A device as set forth in claim 11 characterized in that the microspectrometer is arranged in such a way that spectral resolution of at least fifteen wave numbers is achieved.

13. A device as set forth in one of claims 7 through 12 characterized in that the microspectrometer of the optical identification

unit is replaced by other suitable spectroscopic devices in dependence on the particles to be analyzed.

14. A device as set forth in one of claims 7 through 13 characterized in that the control is an electronic control which includes a programmable AD-converter card with integrated processor and an integrated control program.

Abstract

The invention concerns an optical method and an associated device for characterization of particulate systems, with which the particles present in a particulate system, for example a clean room, can be detected in respect of quantity and size and at the same time information about the identity of the particles can be provided. In accordance with the invention an air flow from the ambient air is guided at a defined speed by a particle feeder past a first scattered light measuring unit and the scattered light is detected, the speed of the particle is then reduced and the particle which is moved at the reduced speed in the air flow is identified in an identification unit by means of interaction with monochromatic light.

In accordance with the invention identification of the particles is effected by combined laser-Raman spectroscopy which, with a short exposure time, by virtue of the use of powerful light sources, strong-light optics and in particular by the renunciation of high levels of optical resolution, affords results which can be used for automated evaluation. The low level of spectral resolution is used to advantage.

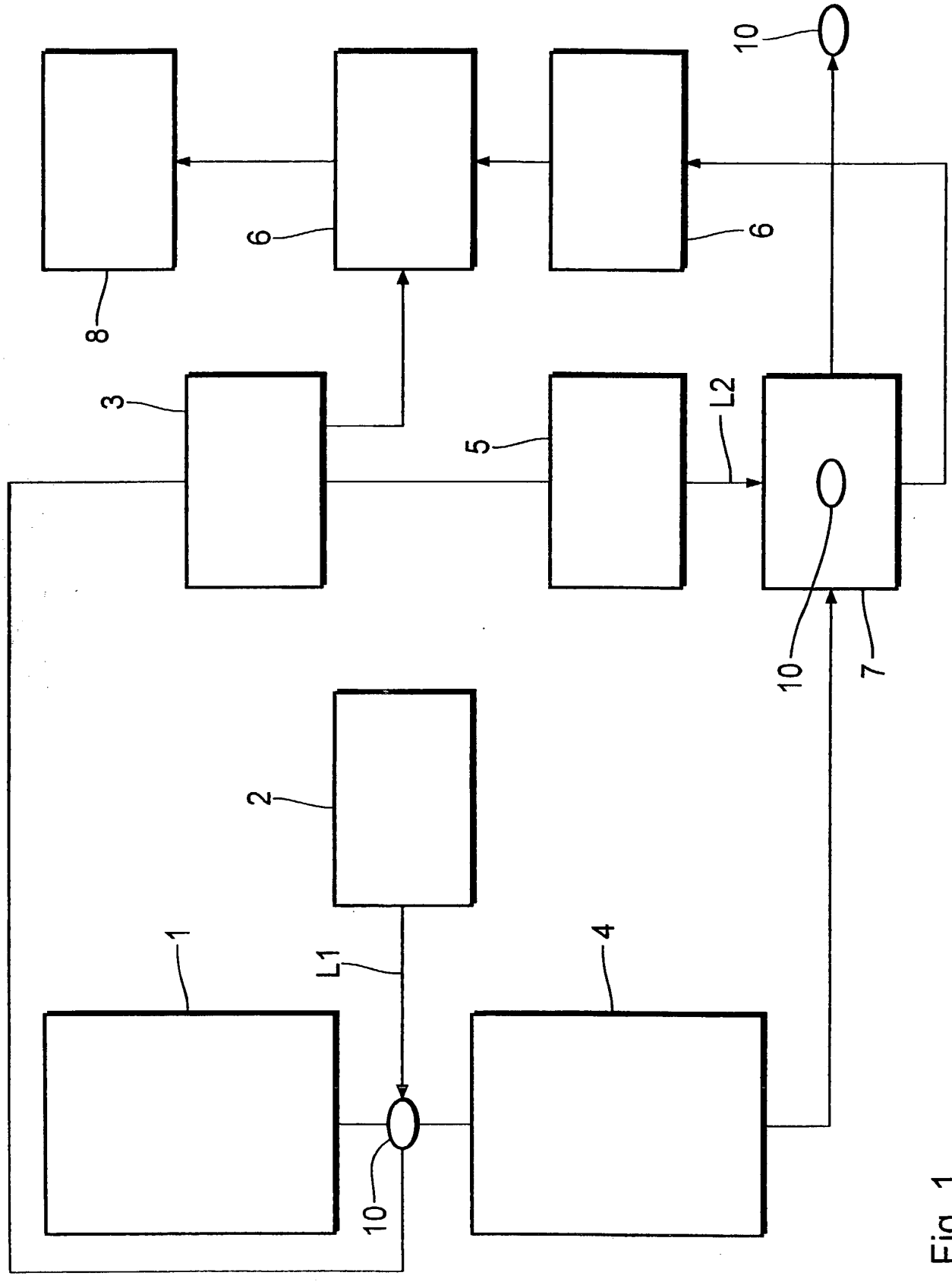


Fig. 1

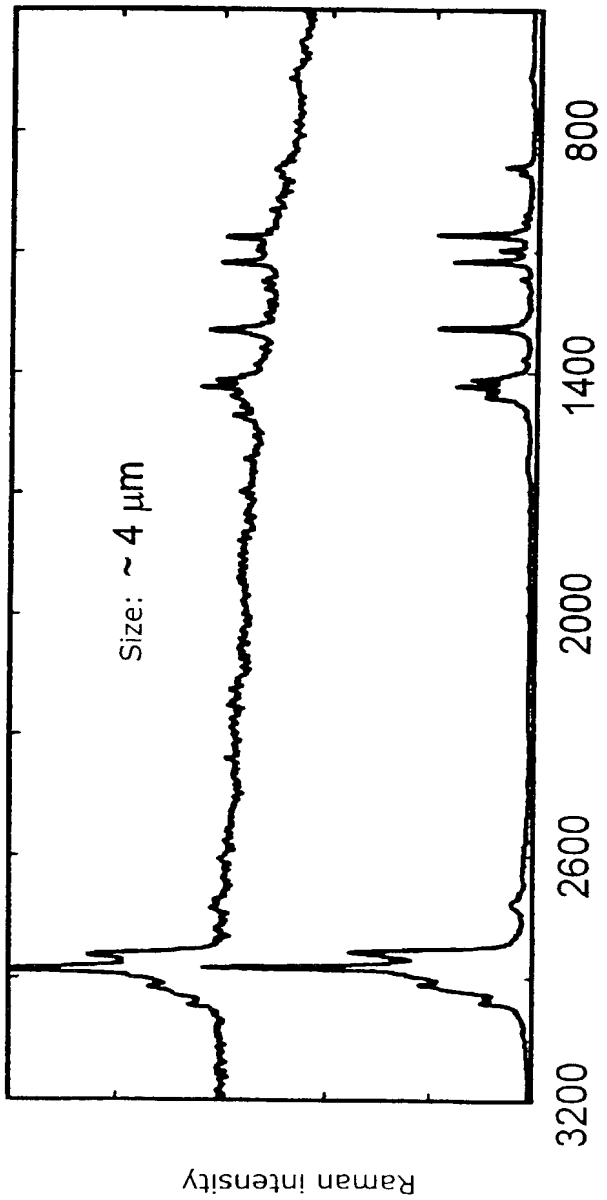

Size: ~ 4 µm
Raman intensity
3200
2600
2000
1400
800
Wave number cm-1

Fig. 2

Practitioner's Docket No. 7040-57 *PATENT*

COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION, OR C-I-P)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is for a national stage of PCT application.

INVENTORSHIP IDENTIFICATION

My residence, post office address and citizenship are as stated below, next to my name. I believe that I am an original, first and joint inventor of the subject matter that is claimed, and for which a patent is sought on the invention entitled:

TITLE OF INVENTION

OPTICAL METHOD FOR CHARACTERIZATION OF PARTICULATE SYSTEMS AND DEVICE FOR CARRYING OUT SAID METHOD

SPECIFICATION IDENTIFICATION

The specification was described and claimed in PCT International Application No. EP00/07937 filed on August 17, 2000 .

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, Section 1.56, and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent .

PRIORITY CLAIM (35 U.S.C. Section 119(a)-(d))

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

Such applications have been filed as follows.

PRIOR FOREIGN APPLICATION(S) FILED WITHIN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS APPLICATION AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. SECTION 119(a)-(d)

COUNTRY	APPLICATION NUMBER	DATE OF FILING DAY, MONTH, YEAR	PRIORITY CLAIMED UNDER 35 U.S.C. SECTION 119
Germany	199 46 110.4	17 September 1999	yes

POWER OF ATTORNEY

I hereby appoint the following practitioner(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

APPOINTED PRACTITIONER(S)	REGISTRATION NUMBER(S)
Stephen L. Grant	33390
Eryn R. Ace	44491
Alexander D. Bommarito	44036
Robert J. Clark	45835
R. Eric Gaum	39199
Michael H. Minns	31985
Edwin W. Oldham	22003
Scott M. Oldham	32712
Mark A. Watkins	33813

9

I hereby appoint the practitioner(s) associated with the Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

SEND CORRESPONDENCE TO

Stephen L. Grant
1225 W. Market St.
Akron, OH 44313
USA

Customer Number 021324

DIRECT TELEPHONE CALLS TO:

Stephen L. Grant
330-864-5550

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURE(S)

1-00 Oliver K. VALET
Inventor's signature _____
Date 20.3.2002 **Country of Citizenship** Germany
Residence Berlin Germany DEX
Post Office Address Boxhagener Strasse 86, Berlin D-10245 Germany

2-00 Peter A. GUENTHER
Inventor's signature _____
Date 20.3.2002 **Country of Citizenship** Germany
Residence Berlin Germany DEX
Post Office Address Albrechtstrasse 118, Berlin D-12167 Germany